
Tunable Control of Hydrogel Microstructure by Kinetic Competition between Self-Assembly and Crosslinking of Elastin-like Proteins.

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Public Summary:

The fabrication of three dimensional "bead-string" microstructured hydrogels is rationally achieved by controlling the relative timing of chemical crosslinking and physical self-assembly processes of an engineered protein. To demonstrate this strategy, an elastin-like protein (ELP) amino acid sequence was selected to enable site-specific chemical crosslinking and thermoresponsive physical self-assembly. This method allows the tuning of material microstructures without altering the ELP amino acid sequence but simply through controlling the chemical crosslinking extent before the thermally induced, physical coacervation of ELP. A loosely crosslinked network enables ELP to have greater chain mobility, resulting in phase segregation into larger beads. By contrast, a network with higher crosslinking density has restricted ELP chain mobility, resulting in more localized self-assembly into smaller beads. As a proof of concept application for this facile assembly process, we demonstrate one-pot, simultaneous, dual encapsulation of hydrophilic and hydrophobic model drugs within the microstructured hydrogel and differential release rates of the two drugs from the material.

Scientific Abstract:

The fabrication of three dimensional "bead-string" microstructured hydrogels is rationally achieved by controlling the relative timing of chemical crosslinking and physical self-assembly processes of an engineered protein. To demonstrate this strategy, an elastin-like protein (ELP) amino acid sequence was selected to enable site-specific chemical crosslinking and thermoresponsive physical self-assembly. This method allows the tuning of material microstructures without altering the ELP amino acid sequence but simply through controlling the chemical crosslinking extent before the thermally induced, physical coacervation of ELP. A loosely crosslinked network enables ELP to have greater chain mobility, resulting in phase segregation into larger beads. By contrast, a network with higher crosslinking density has restricted ELP chain mobility, resulting in more localized self-assembly into smaller beads. As a proof of concept application for this facile assembly process, we demonstrate one-pot, simultaneous, dual encapsulation of hydrophilic and hydrophobic model drugs within the microstructured hydrogel and differential release rates of the two drugs from the material.

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